HIGH PRESSURE CARBON BANDS AND THEIR RELATION TO THE SWAN SYSTEM

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ABSTRACT. In this paper the suggestion that the high pressure carbon bands do not form a new electronic system but belong to the $v'=6$ progression of the Swan system is examined in detail. It is shown that available data on these bands do not prove this suggestion. A complete rotational analysis of the bands is expected to decide the point unequivocally and show whether the bands are instances for the proposed phenomenon of inverse induced predissociation.

The high pressure carbon bands were first discovered by Fowler (1910) in the course of his work on the low pressure or Comet-tail bands in tubes containing carbon monoxide at relatively high pressure (100 mm.). The bands rapidly disappeared when the pressure was reduced and they could not be observed in the spark at atmospheric pressure or at a pressure of half an atmosphere with or without capacity. Fowler had obtained these bands along with the Angstrom bands and the Swan bands, though both the latter were very faint. According to him, these bands are double headed and are degraded towards the violet. He did not get these bands in CO_2, petroleum, benzene, CN, or CO mixed with air. These bands were also observed in Borelly's Comet in 1903 and in the Comet of 1910a. The conditions of production of these bands were further investigated by Merton and Johnson (1923). They obtained these bands with greater intensity in condensed discharges in capillary tubes fitted with carbon electrodes. The tubes contained carbon monoxide at pressures of 5 mm. and more. The authors have given a reproduction of these bands photographed under low dispersion. They observed that the Swan bands and the high pressure bands were both present in the light coming from the capillary part of the discharge tube, but the high pressure carbon bands were isolated in the bluish jets where the two ends of the capillary merged into the wider parts of the tube. It was further observed that the introduction of a little CO_2 made the bands disappear which appeared again only after a short while in which time presumably the CO_2 was reduced to monoxide by the carbon electrodes.

Johnson and Asundi (1929) photographed the system in the first order of a 21 ft. grating with a view to doing the rotational analysis of these bands and thereby deciding about their emitter. The discharge tube in their experiment was 60 to 70 cm. long having a bore of about 15 to 20 mm. One of the electrodes at least was made of carbon. The tube was filled with carbon monoxide at a pressure of 20-40 mm. The high pressure carbon bands were strongly developed on passing a condensed discharge from a ½ K. Watt, 15,000

volt transformer. Further observations indicated that impurities such as hydrogen, carbon-dioxide, water vapour inhibited the formation of these high pressure carbon bands and the tube always attained its best condition after running for about an hour and removing meanwhile any little hydrogen through the palladium regulator attached to the discharge tube. The light filling the wide bore of the discharge tube gave an appearance of dense pale blue puffs of smoke which was rich in high pressure carbon bands. These pale blue puffs were threaded with a green narrow ribbon which showed only the Swan system. By the proper selection of plates and careful search on them the authors were able to record six more bands which along with the previously recorded bands formed one v'' progression ($v'=0$). They represented the bands by the formula

$$\nu = 29212 - (1627v'' - 11.7v''^2) \quad \ldots \quad (1)$$

Table (I) contains the list of bands observed together with their visually estimated intensity and the v'' numbering as proposed by them. The (O−C) values are given in the last column.

TABLE I

v''	Intensity	λ air I.A.	ν vac.	O−C
0	1	3419	29241	+29
1	1	3619.5	27620	−23
2	1	confused	—	—
3	2	4093	24426	−10
4	7	4368.8	22833	− 8
5	15	4680.2	21361	− 8
6	1	V faint	—	—
7	5	5434.9	18394	− 2
8	10	5899 3	16946	+ 2
9	8	6442.3	15518	+ 1
10	6	7083 2	14114	+ 2
11	4	7852.5	12731	0

The close agreement between the coefficients of v'' of this equation and those of v'' in the equation representing the origins of the Swan bands led them to conclude for the first time that the final state of both these systems is the same, the accepted equation for the Swan system representing the origins being

$$\nu = 19379.20 + (1773.42v' - 19.35v'^2) - (1620.28v'' - 11.67v''^2).$$

Thus the two band systems were shown to be due to a common emitter. This emitter is now accepted to be the C_2 molecule on various and sound grounds both theoretical and experimental. The common final level in these two band systems was considered to be the normal state of the C_2 molecule. This has been confirmed lately by absorption experiments (Klemenc and others, 1934). The small difference between the coefficient of v'' in the two equations is well accounted for by the fact that in the equation for the high pressure

carbon bands the data on heads have been used while in that for the Swan bands those on the origins are used. The apparent double character of the high pressure bands as seen under low dispersion is due to the condensations which constitute the P and R branches. The high pressure carbon bands then form a band system which consists of only one v'' progression. Numerous examples are, however, known of band systems having only one progression. Even in the same molecule, as for example in CO, we have the Herzberg-Asundi bands and 3—A bands involving different electronic states but having only one progression.

Fox and Herzberg (1937) and later Herzberg (1939) discussing the processes of dissociation, predissociation, and recombination refer to the C_2 molecule. In a three body recombination the energy of formation may be carried away as excitation energy by one or both of the resulting systems. Thus in discharges in CO under high pressure the two carbon atoms formed by dissociation combine to form C_2 while the energy of formation of C_2 may be directly utilised in exciting it. This energy is identified by Herzberg with the excitation energy of the high pressure carbon bands. The process is according to him one of chemiluminescence and the observation of Johnson and Asundi (1929), that the high pressure carbon bands are completely isolated in the smoky puffs outside the discharge while in the path of the discharge itself the bands are absent, support this view. If this is correct, as it seems likely, the lower limit of dissociation energy of C_2 is accurately determined as 3.6 volts. Herzberg, however, goes one step further and suggests that the high pressure carbon bands do not involve a new electronic level but are extensions of the Swan system and form indeed the $v'=6$ progression. If this view is correct, a serious difficulty arises in visualising the process outlined above. One can understand the excitation of $v'=6$ progression in C_2 on the basis of the availability of energy of 3.6 volts, but it is rather surprising that the excitation should exhibit such great selectivity that practically none of the lower vibrational levels are excited. If it is true that the high pressure carbon bands are really extensions of the Swan system, this great selectivity of excitation can only be reconciled by the introduction of a new phenomenon the so-called inverse induced predissociation. Thus according to Herzberg, the two carbon atoms which form C_2 recombine only when they approach each other on a potential curve from which a radiationless transition into the upper state of the Swan bands is possible under the action of a third particle. Once formed the molecule will not predissociate again except after collision (induced predissociation). No other example of such inverse induced predissociation has, however, so far been observed. The observation that the bands are very strong in the glowing cloud surrounding the main path of the discharge in the experimental tube is taken as evidence of the chemiluminescent nature of exciatation brought about according to this mechanism. Asundi and Pant (1942), however, have found in spectra of gases excited by high frequency discharge only these high pressure carbon bands to the exclusion of all other bands of C_2 including those of the Swan system

obviously in the path of direct discharge. Before the concept of inverse induced predissociation is invoked, it becomes, therefore, necessary to investigate in detail the experimental evidence for regarding the high pressure carbon bands as extensions of the Swan system. As far as we are aware, no definite evidence has been adduced to prove that the high pressure carbon bands are extensions of the Swan system. Unless this is proved unequivocally, there is no need for the conception of inverse induced predissociation and the value $D(C_2) = 3.6$ volts will have to be regarded only as the lower limit.

One argument in support of the view that the high pressure carbon bands are extensions of the Swan system may be that these bands appear to fit in the (v' v'') scheme for the Swan bands forming the $v'=6$ progression if the following formula* for the heads is used :

$$\nu_h = 19354.9 + (1765.3v' - 16.35v'^2 - 9v'^3) - (1625.9v'' - 10.95v''^2) + 1.8v'\,v'' \quad \ldots \quad (III)$$

This evidence is collected in Table 2 in which the wave numbers of the band heads, as recorded by Johnson 1927 for the Swan system and by Johnson and Asundi (*loc. cit.*) for the high pressure system, are given alongside the wave numbers (in italics), calculated from the above equation. The agreement between the observed and calculated values is indeed remarkable. This, however, is mainly brought about by the last term, $1.8v'\,v''$, in the equation. Thus if the values are calculated ignoring that term, we shall find that the agreement between the observed and the calculated values which is as good as one can expect from an equation representing the heads for bands of low quantum numbers, becomes increasingly worse for higher quantum numbers. These data are collected in Table 3. The theoretical significance of the term $1.8\,v'\,v''$, is not very clear and in earlier literature this term has been used to bring greater agreement between the observed and the calculated values when band head data were to be expressed by means of a formula. Evidently it serves to take into account the varying distance between the head and the origin of a band and also the irregularities in the vibrational levels brought about by various causes such as perturbations.† It is not possible to separate the contributions of these two effects by this term alone. The fact that the (O−C) values become unusually great for higer quantum numbers in this special case seems to indicate the existence of strong irregularities in the spacings of the vibrational levels. This is further confirmed as we shall see presently when we discuss the data in the light of the equation for the origins of the bands of the system.

Generally the ν_h equation is not found to be truly representing a band system; further in some cases because of ill-defined heads, the measurements may not be very accurate. If, however, it is possible to do fine structure

* ' Report on Band Spectra of Diatomic Molecules ' by W. Jevons, pp. 11, 61 and 62.

† The cubic term in v' in eqn. III would apparently indicate perturbations. Such a term, however, is not needed if origins are used, *e.g.*, eqn. II.

analysis of the bands of the system then the system can be truly represented by ν_0 equation. Fine structure analysis of each band gives ν_0 and we can thus find $(\nu_0 - \nu_h)$ experimentally for each band. This value can also be calculated theoretically by the equation :

$$\nu_0 - \nu_h = \frac{(B' + B'')^2}{4(B' - B'')} \div \frac{(B'_0 - \alpha'v' + B''_0 - \alpha''v'')^2}{4(B'_0 - \alpha'v' - B_0'' + \alpha''v'')} \qquad \text{(IV)}$$

where B_0, α, and v, have their usual significance. The variation of ν_h from ν_0, from band to band is of great importance in checking the analysis. It is easy to deduce from the above expression that for the bands degraded towards the red ($B' < B''$) the interval increases with diminishing v' (v'' constant) and also with increasing v'' (v' constant) provided $3B' > B''$ and for the bands degraded towards the violet ($B' > B''$) the interval increases with increasing v' (v'' constant) and also with decreasing v'' (v' constant) provided $B' < 3B''$. The Swan bands and the high pressure bands are degraded towards the violet and so the variation in the interval must increase regularly with increasing v' (v'' constant) and also with diminishing v'' (v' constant). Table 4 gives the intervals. In italics are given the theoretically calculated intervals (Eqn. IV) and in normal type the intervals obtained from the origin equation and the head data given by Johnson (1927) for the Swan bands and Johnson and Asundi (*loc. cit.*) for the high pressure carbon bands. The values for origins and heads are given in Table 5. For such of the bands marked ^{h}, ν_0 values directly observed are available and these are exactly the same as those calculated by the equation. ν_0 values for some of the bands of the high pressure carbon system can be determined approximately by the data given by Johnson and Asundi. These are, however, discussed later. The values of ν_0 used in the table are those calculated by Johnson's equation. It is seen from Table 4 that the interval regularly decreases as it should, in going from the (0, 0) band to higher v'' values for a fixed v'. The (O − C) values for the intervals (Table 6) also indicate this regularity and are negligibly small up to and including bands with $v'=4$. Indeed, the average (O − C) of about 3.8 cm^{-1} for the four bands (0, 0), (0, 1), (1, 1), and (1, 0) for which the origin data of Shea (1927) are available, will practically vanish if Shea's data are used. They are, however, large for bands with $v'=5$ and are very much more so for the bands of the high pressure system which are regarded as forming the $v'=6$ progression of the Swan system. It may be further remarked that the bands with $v'=5$ have very low intensity so that the band head data on them will not have the same accuracy as the rest and high (O − C) values may, in part, be due to this cause.

This discussion indicates either that the high pressure carbon bands cannot be regarded as extensions of the Swan system or that violent perturbation occurs far bands with $v'=5$ and 6. Since the fine structure analysis of the bands having $v'=5$ has not been done, it is not possible to decide between these two alternatives. There is also no other electronic level known in C_2 which can bring about this perturbation. Hence it is not possible to prove conclusively by this evidence alone that the high pressure carbon bands are extensions

of the Swan system. It should also be mentioned that the two bands with $v'=5$ recorded by Johnson may not be genuine bands at all but chance accumulation of structure lines of bands preceding them in the sequence. Shea (*loc. cit.*) records only one of them *viz.*, (5, 4) band. If, therefore, these two bands are neglected the Swan band system definitely ends with $v'=4$ and hence the high pressure carbon bands have to be regarded as forming an independent system.

The second argument which appears to favour the view that the high pressure carbon bands are extensions of the Swan system is probably the intensity distribution. Thus if the Condon parabola for the Swan system be extended to higher quantum numbers, it appears to pass through bands which are relatively more intense in the high pressure system. It must be remembered, however, that in the first place, the intensities recorded are photographic intensities visually estimated only and are to be regarded as descriptive rather than quantitative. Secondly, all the bands are not recorded under identical conditions of excitation and on one plate. Johnson and Asundi have already drawn attention to the main features of the peculiar intensity distribution in the high pressure carbon band system which has a notable minimum at about the (0, 6) band. Similar oscillating intensity phenomena are observed in certain other band systems (Johnson and Jenkins, 1926). Therefore this evidence from the intensity distribution can at best be used only as a subsidiary evidence.

We shall now discuss the experimental evidence in favour of the view that the high pressure carbon bands form an independent system :—

(*a*) These bands are obtained not only in the smoky puffs outside the path of the discharge as in Johnson and Asundi's experiment (1929) but also in the direct path of the discharge as recorded by Asundi and Pant (1942). This means that chemiluminescence is not the only mode of excitation which will bring up these bands. Asundi and Pant (1942) have observed only the high pressure carbon bands in their experiments on high frequency discharge to the exclusion of all other band systems due to C_2.

(*b*) Although a complete analysis of the rotation structure of the high pressure carbon bands has not been done, the fine structure data recorded by Johnson and Asundi (*loc. cit.*) allow us to extrapolate the origins of the four bands for which the data are recorded. The origins thus extrapolated can at least be regarded as upper limits. This allows us to find out the lower limit of the distance between the head and the origin in these bands. The values thus obtained are given in column 4, Table 7, and can be compared with the values of $(\nu_0-\nu_h)$ given in column 5 calculated as before by equation IV, assuming the high pressure carbon bands as extension of the Swan system. In column 6 are tabulated the values of the doublet separation of the high pressure carbon bands as observed by Fowler (*loc. cit.*). These double heads as already pointed out by Johnson and Asundi represent the P and R heads. The data of Fowler are admittedly inaccurate but it is seen that the values given in column 6 are of comparable magnitude if we keep in view the two limitations, *viz.*, that the data

TABLE II

v' \ v''	0	1	2	3	4	5	6	7	8	9	10	11
0	19355.0 *19354.90*	17740 *17739 95*	16148 *16146.95*									
1	21105 *21102 95*	19490 *19490*	17899 *17899*	16339 *16339*								
2	22812 *22812.9*	21202 *21202*	19612 *19612*	18043 *18044.6*	16498 *16498.9*	14972 *14975*						
3		22870 *22869*	21282 *21282*		18171 *18172.55*	16649 *16650.6*	15149 *15150.6*					
4			22902 *22903*	21343 *21339.4*		18276 *18277.2*	16778 *16778 9*	15301 *15302 4*				
5					21368 *21367.8*			16875 *16875 4*				
6	29241 *29163.7*	27620 *27559.6*	... *25977*	24426 *24416*	22883 *22875 5*	21561 *21561.9*	19871 *19867*	18394 *18394.6*	16946 *16943 7*	15518 *15514.5*	14114 *14107.7*	12731 *12722.6*

In italics are given the calculated values according to equation

$$\nu_h = 19354.9 + (1765.3v' - 16.35v'^2 - .9v'^3) - (1625.9v'' - 10\ 95v''^2) + 1.8v'v''.$$

In normal type are given the values (correct to 5 figures) observed by Johnson for the Swan bands up to $v'=5$, and for $v'=6$ those observed by Johnson and Asundi for the high pressure carbon bands.

TABLE III

v' \ v''	0	1	2	3	4	5	6	7	8	9	10	11
0	19355.0 *19354.9*	17749 *17739.9*	16148 *16146.95*									
1	21165 *21162.65*	19496 *19485.5*	17899 *17895.0*	16330 *16324.4*								
2	*22812 *22812.9*	21202 *21195.4*	19612 *19604.2*	18043 *18032.5*	16468 *16453.9*	14972 *14954*						
3	22879 *22864.0*	21282 *21270.7*		18171 *18148.9*	16549 *16521.5*	15149 *15116*						
4		22992 *22887.5*	21340 *21315.2*		18276 *18239.5*	16778 *16734.5*	15311 *15250.7*					
5				21368 *21332.0*			16878 *16814*					
6	29241 *29163.7*	27620 *27548*	... *25955.4*	24426 *24373.6*	22883 *22835.3*	21361 *21307.0*	19871 *19803.2*	18394 *18310.0*	16946 *16857.3*	15518 *15417.6*	14114 *13999.7*	12731 *12603.8*

In italics are the calculated values after being corrected for the term $1.8\,v'\,v''$, i.e., according to equation

$$\nu_{\text{obs}} = 19354.9 + (1765.3v' - 16.35v'^2 - .5v'^3) - (1625.9v'' - 10.95v''^2).$$

In normal type are the values as given in Table II observed by Johnson and Johnson and Asundi.

TABLE IV

v' \ v''	0	1	2	3	4	5	6	7	8	9	10	11
0	18.94 *23.06*	16.0 *20.4*	13·3 *18.25*									
1	23.6 *26.55*	19.3 *23.08*	16.2 *20.36*	13·4 *18 19*								
2	31.8 *31.39*	23.0 *20.64*	18.8 *23 09*	15 3 *20.33*	12 6 *18.13*	13.8 *16 32*						
3		32·5 *31.64*	25·4 *26.74*		16.7 *20 30*	13.8 *18.01*	11·5 *16.24*					
4			42·7 *31.91*	33·5 *29.56*		24.8 *20 25*	21·3 *18 00*	19 4 *16 16*				
5					56.5 *26.99*			42·4 *17.85*				
6	76.8 *135.85*	79.6 *76.63*		107.2 *40.41*	102.0 *32.55*	99.1 *27.12*	87.1 *23.21*	86.5 *20 22*	79.6 *17.55*	76.1 *16 00*	72.0	69.0

Figs. in italics are calculated values.

Figs. in normal type are those obtained from difference of figures in Table V.

TABLE V

v' \ v''	0	1	2	3	4	5	6	7	8	9	10	11
0	19354.96 *19373.9**	17739.7 *17755.7**	16147.5 *16160.8**									
1	21194.3 *21127.9**	19490.4 *19509.7**	17898.7 *17914.9**	16329.9 *16343.3**								
2	22811.5 *22843.3*	21202.1 *21225.1*	19611.4 *19630.2*	18043.4 *18058.7*	16497.9 *16510.5*	14971.9 *14985.7*						
3		22869.3 *22901.8*	21281.5 *21306.9*		18170.5 *18187.2**	16648.5 *16662.3**	15149.3 *15160.8*					
4			22902.2 *22944.9*	21339.8 *21373.3*		18275.3 *18300.3*	16777.5 *16798.8*	15301.2 *15320.6*				
5					21367.9 *21424.4*			16877.5 *16919.9*				
6	29241 *29317.8*	27620 *27699.6*	... *26104.7*	24426 *24533.2*	22883 *22985.0*	21361 *21460.1*	19871 *19958.6*	18394 *18480.5*	16946 *17025.6*	15518 *15594.1*	14114 *14186.0*	12731 *12799.98*

ν_0 calculated by the Johnson equation (in italics) $\nu_0 = 19373.87 + (1773.42v' - 19.35v'^2) - (1629.88v'' - 11.67v''^2)$

ν_h observed by Johnson for the Swan bands and by Johnson and Asundi for the high pressure carbon bands (in normal type).

* Denotes that the value is directly observed.

TABLE VI

O−C Values of Intervals

v′ \ v″	0	1	2	3	4	5	6	7	8	9
0	−4.12	−4.14	−4.95							
1	−2.95	−3.78	−4.16	−4.79						
2	+.41	−3.64	−4.29	−5.03	−3.53	−2.52				
3		+.86	−1.34		−3.60	−4.26	−4.74			
4			+10.79	+6.64		+4.55	+3.30	+3.24		
5					+29.52			+24.45		
6	−59.03	+2.97		+66.79	+69.47	+71.98	+64.27	+66.27	+61.72	+60.1

are not accurate and the difference represents the distance between the P and R heads and not that between the P head and the origin which will be the greater.

TABLE VII

v''	ν_h	ν_0 observed	$(\nu_0-\nu_h)$ observed	$(\nu_0-\nu_h)$ calculated by equation IV	Doublet width Fowler (1910)
4	22883	22934	51	32 53	64
5	21361	21410	49	27.12	73
8	16976	16976	30	17 88	55
9	15518	15545	27	16 00	51

This table clearly shows that the moment of inertia of the molecule emitting the high pressure carbon bands is not correctly given if the bands are regarded as extensions of the Swan system.

(*c*) There is yet another experimental evidence which is fully discussed by Johnson and Asundi (*loc. cit.*) and which also favours the view that the high pressure carbon bands are to be regarded as a separate system. This concerns the triplet electronic interval. It should be remembered that the high pressure carbon bands are due to a transition between two triplet II levels, the lower of which is, without doubt, the same as that of the Swan system. If, therefore, the high pressure carbon bands did not involve a new electronic initial level, the triplet width should be the same as that for the Swan bands. It has been observed, however, that the triplet width in high pressure carbon bands is greater than that of the Swan bands. This is in perfect agreement with the view that the initial electronic level of the high pressure carbon bands is not the same as that of the Swan system but lies higher. Since the triplet widths represent the difference between the triplet separations of the initial and the final states, the fact that the triplet width in the high pressure carbon bands is greater than in the Swan bands, is a consequence of the regular diminution in triplet width for the higher excited $^3\Pi$ level of the high pressure carbon bands,

Thus we see that a certain vibrational extrapolation of the Swan bands can include in that system the high pressure carbon bands also. The peculiar intensity distribution may also to a certain extent support the inclusion of the high pressure bands in the Swan system but there are serious difficulties for this view. One is that the $(\nu_0-\nu_h)$ values for the high pressure carbon bands calculated on this assumption do not agree with the values which can be deduced from the observed data. This will then mean that we shall have to assume the existence of a strong perturbation of the $v'=6$ level of the Swan system. Such a perturbation can be brought about by the vibrational terms of another electronic level which possesses the right properties for such a perturbation. No such electronic level has been so far observed in the C_2 molecule. A study of the possible potential curves of the C_2 molecules indicates that the

existence of such a perturbing electronic state possessing vibrational levels is not very probable. On the other hand ($v_0 v_h$) values as well as the increased interval between the triplet components of the high pressure carbon bands as compared to the Swan bands indicate that the high pressure carbon bands are to be regarded as an independent system. The controversy can, therefore, only be resolved by more complete data on the rotational structure of the high pressure carbon bands. In view of this and of the fact that the high pressure carbon bands can be excited by methods which are not necessarily chemiluminescent in nature, has to be reserved as to the existence of the phenomenon of inverse induced predissociation which is necessary to explain the great selectivity of the bands, if they are not an independent system but only extensions of the Swan band system.

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